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HOT ISOSTATIC COMPACTION OF TUNGSTEN - URANIUM DIOXIDE FUELS WITH HIGH-VOLUME FRACTION OF URANIUM DIOXIDE

by Paul F. Sikora and Andrew C. Millunzi

Lewis Research Center
Cleveland, Ohio

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SUMMARY

Dense (>98 percent of theoretical) tungsten - uranium dioxide cermets containing 30 to 70 volume percent uranium dioxide were fabricated by hot isostatic compaction of tungsten-coated uranium dioxide particles. Both thin plates (0.020-in. or 0.051-cm thick) and small-diameter rods (0.20-in. or 0.51-cm diam) were successfully produced at compaction conditions of 3100° F (1700° C) and 30 000 psi (207 MN/m^2). In addition to volume-fraction loading of uranium dioxide, other variables studied in this program were the effect of (a) fuel particle size in the range of 50- to 110-micron diameter, (b) molybdenum or a molybdenum alloy (TZM) in container assemblies, and (c) compaction schedule and pressure (up to 42 000 psi or 295 MN/m^2).

Fully dense cermets were produced with all combinations of the range of variables studied. The surface smoothness of the cermets was affected only by the choice of container materials. Containers produced from TZM resulted in smooth cermet surfaces; however, unalloyed molybdenum containers often resulted in irregular surfaces.

The feasibility of applying a thin tungsten cladding to the cermet surfaces as an integral part of the compaction process was also demonstrated in this study.

Comparison of these hot isostatically compacted cermets with similar cermets produced by hot-roll compaction indicated that less deformation of the fuel particles resulted from hot isostatic compaction.

INTRODUCTION

Nuclear power generation systems for space applications are being studied at the Lewis Research Center. In order to minimize shielding requirements and powerplant

weight, these systems probably will use fast reactors that contain a high-volume fraction of fully enriched fuels, will use liquid-metal coolants, and will operate at temperatures above 1800° F (980° C). Dispersion-type fuel elements containing high volume fractions of uranium dioxide particles in a refractory metal matrix and clad with a refractory metal are prime candidates for these cores. Tungsten and tungsten alloys are prime matrix and cladding material candidates because of their high-temperature strength and thermal conductivity.

General requirements for cermet fuel compacts are high density (>98 percent of theoretical) and uniform fuel distribution with complete separation of the individual fuel particles. These requirements are needed for maximum cermet strength and uniform heat generation. Another requirement for these cermets is smooth surfaces. Smooth surfaces are needed on these cermet fuels to provide intimate contact between the fuel compact and the surface claddings for maximum heat transfer to the reactor coolant.

In previous studies (refs. 1 and 2) three fabrication techniques were investigated for the production of fully dense tungsten - uranium dioxide (W-UO_2) cermets with low volume fractions of UO_2 (up to 35 vol. percent UO_2). These techniques all involved canning W-coated UO_2 particles in a suitably ductile can material and applying heat and pressure by various means. The use of the precoated particles assured the attainment of uniform fuel distribution and fuel particle separation. These techniques were

(1) Hot isostatic compaction: powder compaction achieved by pressurized inert gas acting on a simultaneously heated sample for an extended time (several hours).

(2) Hot pneumatic impaction: powder compaction achieved by impacting a heated sample with a very short-duration pressure pulse in a high-energy-rate forming machine.

(3) Hot-roll compaction: powder compaction achieved by rapidly rolling a heated sample in a conventional metal-working rolling mill.

After comparing the results of the previous studies (refs. 1 and 2), we decided that hot isostatic compaction offered the best potential for producing W-UO_2 cermets containing up to 70 volume percent UO_2. Therefore, the purpose of the present study was to develop further the hot isostatic compaction method for fabricating W-UO_2 composites containing a high volume fraction of fuel (up to 70 vol. percent UO_2).

Presented herein are the results of a study conducted on the effects of selected variables on the fabrication of dense W-UO_2 flat plates and rods by the hot isostatic compaction process. These variables include volume fraction of UO_2 particle loading, fuel particle size, container material, and compaction schedule and pressure. In addition, a limited study was made to determine the feasibility of applying a tungsten cladding during the compaction of the cermets. A comparison is made between cermets produced by roll compaction and hot isostatic compaction. Density measurements and metallographic examinations were used to evaluate each of the test parameters.

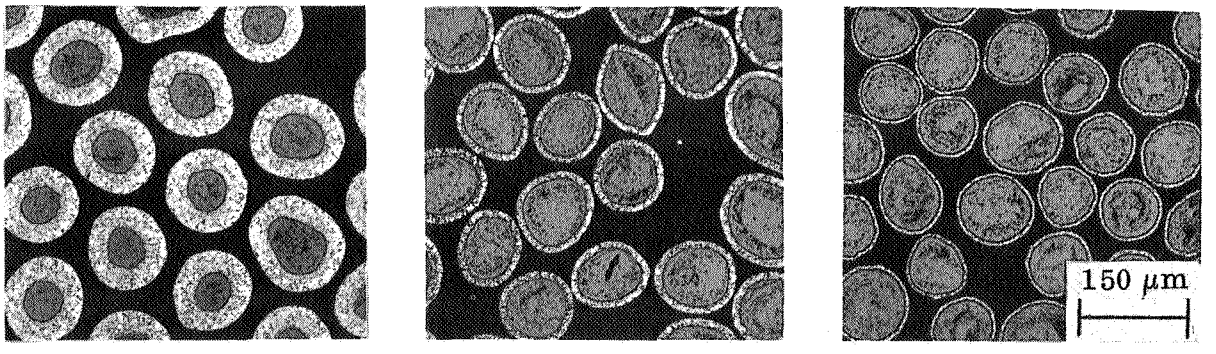
EXPERIMENTAL MATERIALS AND PROCEDURES

In general, the hot isostatic compaction process consists of consolidating powders to a desired configuration by the following procedure: (a) Loose powders are poured into a shaped container, and the container is sealed under vacuum. (b) The sealed container is placed in an inert gas autoclave, which applies high pressure and high temperature to the container. The system pressure and container temperature are raised according to a predetermined schedule with a resulting deformation of the container and compaction of the loose powder. (c) The container material is removed by either chemical leaching or mechanical stripping. A more detailed description of the hot isostatic compaction process is reported by Paprocki, Hodge, Boyer, and Getz in reference 3.

The specimen loading, pretreatment, and evaluation described in this report were performed at the Lewis Research Center. The compaction runs were conducted at the Columbus laboratories of Battelle Memorial Institute under a NASA contract.

Materials

Commercially prepared, spherical, depleted UO_2 fuel particles were used in this study. The average size of the UO_2 particles was varied from 50 to 110 microns. The high-purity UO_2 particles (99.9 percent minimum purity) were precoated with tungsten by vapor deposition in a fluidized bed with the use of the hydrogen reduction of tungsten halides (ref. 2). Most of the work was done with particles containing 30, 50, or 70 vol-



(a) Tungsten - 30 volume percent uranium dioxide.

(b) Tungsten - 50 volume percent uranium dioxide.

(c) Tungsten - 70 volume percent uranium dioxide.

Figure 1. - Photomicrographs of typical tungsten-coated uranium dioxide particles. Etchant, Murakami's reagent.

ume percent UO_2. Photomicrographs of typical coated particles of these three fuel loadings are shown in figure 1. Typical analyses of the tungsten-coated particles used in this study are presented in table I.

TABLE I. - CHARACTERIZATION OF TUNGSTEN-COATED URANIUM DIOXIDE PARTICLES

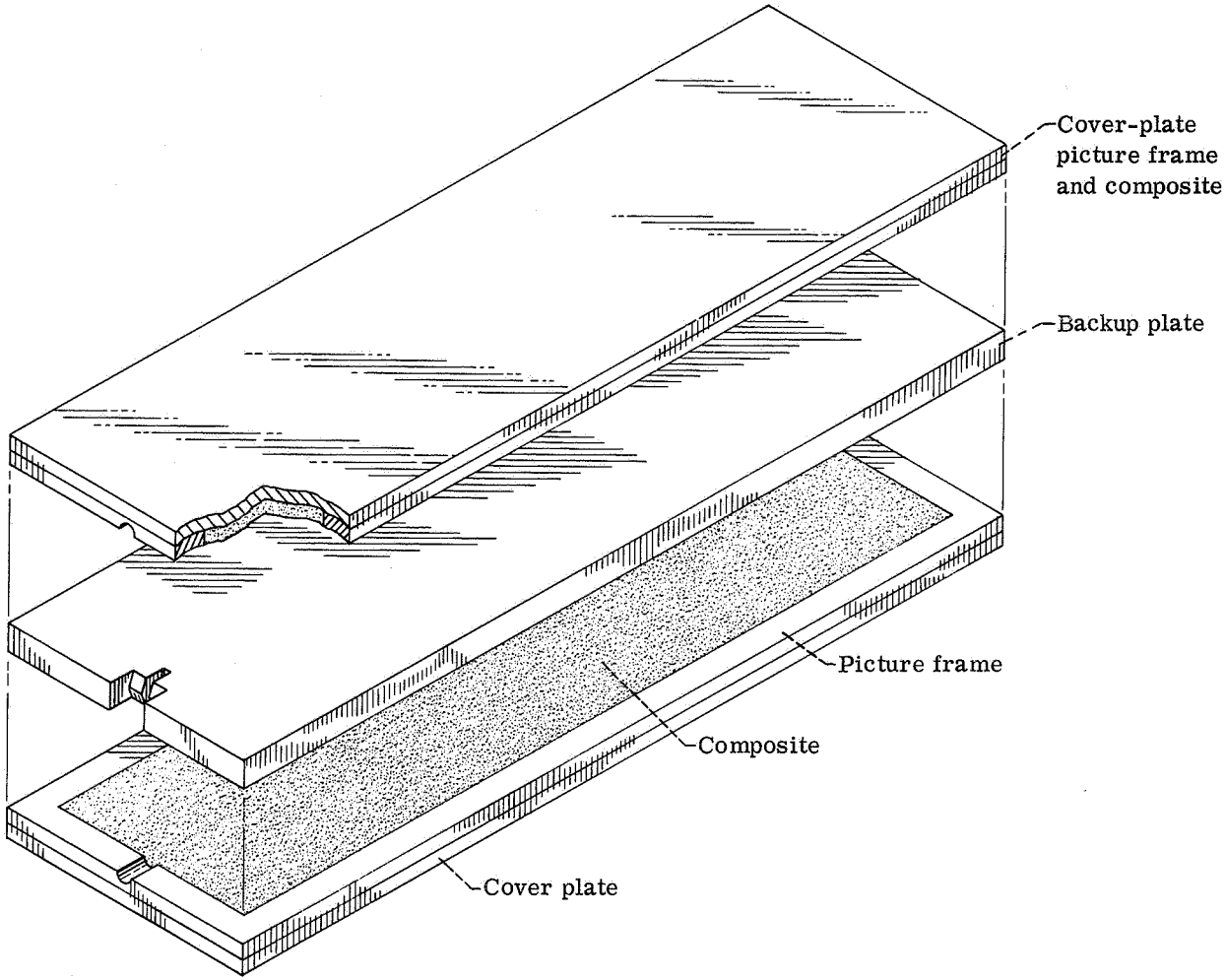
Nominal fuel loading, vol. % UO_2	Average UO_2 particle size, μm	Average tungsten coating thickness, μm	Tungsten, wt %	UO_2, wt %
30	110	28	81.66	18.34
35	50	10	78.12	21.88
50	56	7	63.03	36.80
50	110	15	64.14	35.85
70	110	7	42.68	57.32

The container materials used were molybdenum and a molybdenum alloy, TZM (Mo-0.5Ti-0.08Zr-0.03C). All container materials were commercially produced to sheet form and metallurgical grade purity (99.9 percent minimum).

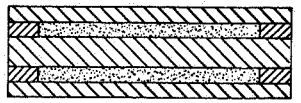
The container assemblies used to fabricate plates are shown in figure 2. The assembly consisted of 0.030-inch- (0.076-cm-) thick molybdenum picture frames and cover plates and a 0.125-inch- (0.318-cm-) thick backup plate shown in figure 2(a). The assembly was considered the "standard" container for flat plates. A cross-sectional view of the standard container is shown in figure 2(b). The container design allowed the compaction of two specimens per container. Typical specimens sizes were 1.25 inch wide by 6 inches long by 0.020 inch thick (3.18 by 15.24 by 0.0051 cm). Several variations of this design (figs. 2(c) to (e)) were used. These variations evolved from the efforts to produce specimens with smooth surfaces and to tungsten-clad the cermet surfaces.

The molybdenum backup plate used in the plate-type specimen (fig. 2(a)) and the TZM material were from arc-melted-and-rolled stock. However, the molybdenum used for cover plates and picture frames and the tungsten-foil liners used were from sintered and rolled stock. Vapor-deposited-tungsten liners also were used. These liners were deposited on the internal surfaces of the molybdenum components (fig. 2(c)) by thermal decomposition of tungsten hexachloride (WCl_6).

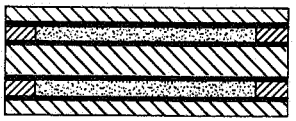
Simple rod configurations (0.2-in. diam by 6.0 in. long or 0.51 by 15.24 cm) were also produced with thin-walled tubes used as containers. These tube-type containers utilized commercially extruded and drawn molybdenum tubing.



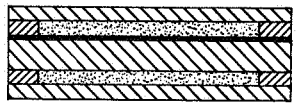
(a) Exploded view of flat-plate container (two plates per container).



(b) Cross-sectional view of standard molybdenum or TZM container design.



(c) Cross-sectional view of tungsten foil or vapor-deposited tungsten on molybdenum.



(d) Cross-sectional view of molybdenum cover plate container design.

(e) Cross-sectional view of another molybdenum or TZM container design.

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Figure 2. - Container designs for flat-plate specimen fabrication by hot isostatic compaction.

Procedure

Because spherical fuel particles are free flowing, much like a liquid, the container could be filled by pouring the particles through a hypodermic syringe. Simultaneously, the containers were vibratory packed to a density of approximately 65 percent of theoretical. The filled containers were then cleaned by 1-hour exposure in vacuum and 1-hour in flowing hydrogen, both at 2500° F (1370° C). This cleaning treatment did not adversely affect the UO_2 and was an excellent one for cleaning tungsten.

All the cleaned, powder-filled containers were backfilled with argon prior to removal from the cleaning furnace. The backfilled containers were placed in argon-filled plastic bags and transferred to the welding chamber. After the plastic bag was removed, the containers were evacuated to approximately 10^{-5} torr (1.5×10^{-3} N/m^2). The small fill hole was then sealed by electron-beam welding.

After closure, the containers were placed in a pressure vessel at a pressure of 50 to 75 psi (0.3 to 0.5 MN/m^2) of helium. The containers were removed from the vessel and immersed in alcohol. Bubbling of the alcohol indicated any leaks in the containers. It was possible to salvage some of the containers that exhibited leakage by inserting the loaded container into a tantalum secondary container and repeating the sealing and leak-testing procedures.

Several compaction schedules were used and are shown in figure 3. Schedule A consisted of heating the specimens to 1200° to 1400° F (650° to 750° C) under a helium pressure of 1000 psi (6.9 MN/m^2) and then increasing the temperature to 3100° F (1700° C) and the pressure to 30 000 psi (207 MN/m^2). This temperature and pressure were held for 3 hours. These conditions were considered the "standard" schedule and were chosen on the basis of the results of the hot isostatic compaction study reported in reference 1. The 3100° F (1700° C) compaction temperature was the maximum temperature attainable in the pressure vessel used in these studies, and the 30 000-psi (207-MN/m^2) compaction pressure was previously determined to be the minimum pressure needed to densify W-UO_2 cermets completely.

Schedule B utilized the same maximum pressure and temperature conditions but different rates of applying the pressure and temperature. To determine the effects of higher compaction pressures, schedules C and D were utilized with maximum pressures of 33 000 psi (228 MN/m^2) and 42 000 psi (290 MN/m^2), respectively. All the schedules utilized the same compaction temperature (3100° F or 1700° C), and all but schedule D included a 3-hour hold at the maximum compaction conditions. The shorter hold time (1.5 hr) used in schedule D was due to limitations of the autoclave in maintaining a satisfactory thermal gradient on the containers during processing.

Evaluation specimens were cut from the compacted containers for density determinations and metallographic examinations. The containers were removed from the density

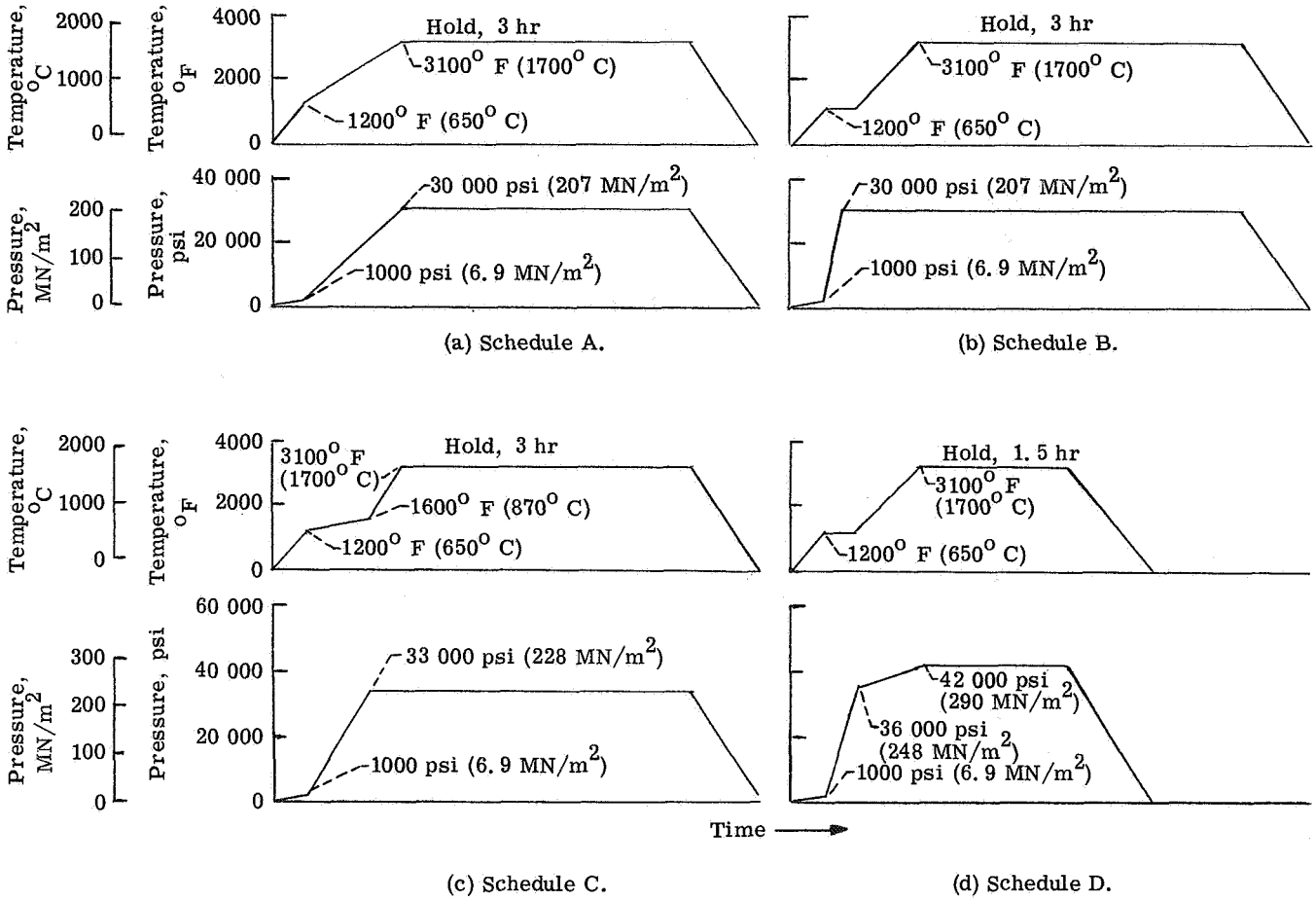


Figure 3. - Hot isostatic compaction schedules used.

specimens by chemical dissolution of the container in a solution of 50 percent nitric acid and 50 percent water. Density measurements were made by the water-displacement technique. Theoretical densities were calculated by using 19.3 grams per cubic centimeter as the density of tungsten and 10.9 grams per cubic centimeter as the density of UO_2. The metallographic specimens were left in the containers to preserve the edges of the cermets.

RESULTS AND DISCUSSION

The effects of volume fraction of UO_2 loading, UO_2 particle size, hot isostatic compaction pressure and schedule, and container material on the density and surface smoothness of W-UO_2 cermets are discussed in the following sections. The materials and compaction conditions are summarized in table II along with the resultant densities of selected cermets. Although all possible combinations of the different variables were not studied, we feel that the number of combinations studied was adequate to illustrate that the variables, within the range investigated, have no adverse effect on the attainment of

TABLE II. - SUMMARY OF TUNGSTEN - URANIUM DIOXIDE SPECIMENS USED TO EVALUATE EFFECTS OF MATERIAL AND COMPACTION VARIABLES

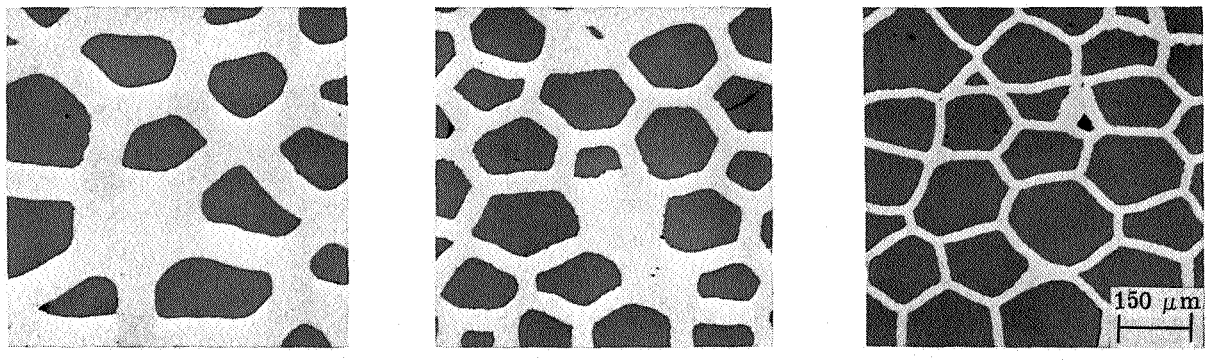
Nominal fuel loading, vol. % UO_2	Average UO_2 particle size, μm	Container		Compaction schedule (see fig. 3)	Density, percent of theoretical	Surface smoothness
		Material	Design given in fig. 2			
Plates						
30	110	Mo	2(b)	C	99.1	Varied
35	50		2(b)	A	97.5	Varied
35	50		2(b)	D	98.4	Good
50	110		2(d)	C	97.5	Varied
70		Mo + W	2(c)	A	100	Good
70		Mo + W	2(e)	A	100	Good
70		TZM	2(b)	A	100	Good
Rods						
30	110	Mo	---	B	100	Poor
50	56		---	B	99.6	
50	110		---	B	99.1	
70	110		---	B	100	

high density and uniform dispersion. The results and discussions presented apply equally to plate and rod specimens because no significant differences were observed in the densities or microstructures of similarly compacted specimens.

Density and Fuel Distribution

Dense cermets (>98 percent of theoretical) were fabricated for the full range and combination of variables studied. Also, uniform particle distribution and complete separation of the fuel particles were achieved under all conditions studied. Typical photomicrographs of cermets containing 30, 50, and 70 volume percent UO_2 are presented in figure 4. No effects of fuel loading or particle size were observed.

As shown in table II, density measurements of two of the specimens were less than 98 percent of theoretical and, apparently, were not fully densified. Subsequent metallographic examination of these specimens revealed that the surfaces were attacked by the leach solution and that some of the surface particles were missing. However, no differences were observed between interior sections of these specimens and those of the es-



(a) Tungsten - 30 volume percent uranium dioxide.

(b) Tungsten - 50 volume percent uranium dioxide.

(c) Tungsten - 70 volume percent uranium dioxide.

Figure 4. - Photomicrographs of fully dense, hot isostatically compacted W-UO_2 specimens. Unetched.

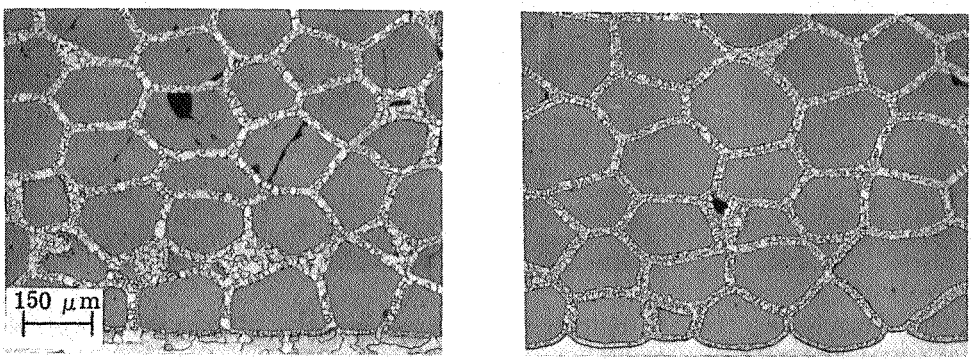
sentially fully dense specimens. Therefore, we feel that all specimens were equally densified during compaction and that the lower density measurements were due to dissolution of the surfaces during the leaching operations. The dissolution was probably caused by alloying between the container and the fuel particle coating during compaction. We believe that this problem can be solved by decreasing the time at temperature and/or changing the cooling conditions after compaction. However, further study of the effects of these changes is needed.

The UO_2 fuel particles were densified during compaction, as can be seen by comparing figures 1 and 4. The UO_2 particles were initially about 94 percent dense, but after compaction they were essentially fully dense. This fuel densification was observed for all loading levels, fuel particle sizes, container materials, and compaction schedules used.

No differences in density or microstructure were observed to result from the two different methods (schedules A and B, figs. 3(a) and (b)) used to attain the standard compaction conditions of 3100° F (1700° C) and 30 000 psi (207 MN/m^2). Similarily, no differences were observed to result from the higher pressures used (schedules C and D, figs. 3(c) and (d)). Thus, we conclude that there is no advantage in using compaction pressures greater than 30 000 psi (207 MN/m^2) at a compaction temperature of 3100° F (1700° C). Also, the method of pressurization is of little importance compared with the maximum compaction conditions achieved.

Surface Smoothness

Surface smoothness (based on metallographic observations) was dependent on the container material used. Smooth surfaces were obtained on the cermets compacted in TZM,



(a) Molybdenum alloy, TZM, cover plate.

(b) Molybdenum cover plate.

Figure 5. - Surface smoothness of W-70 vol. % UO_2 cermets with different container materials. Etchant, Murakami's reagent.

but varying degrees of surface smoothness were found on cermets compacted in unalloyed molybdenum (fig. 5).

During the initial phase of this work, the surfaces of UO_2 particles in contact with the thin sintered-and-rolled molybdenum cover plate were flat and smooth, while the surface particles in contact with the thicker arc-cast-and-rolled molybdenum backup plate were rounded and relatively undeformed. To determine whether this variation in surface smoothness resulted from differences in materials or from the position in the container, two W - 50 volume percent UO_2 cermets were compacted in a specially prepared molybdenum container. For this test, a thin molybdenum cover plate was interposed between the one group of coated particles and the thicker, arc-cast molybdenum backup plate (as illustrated in the container design shown in fig. 2(d)). Cross sections of the resultant specimens are shown in figure 6. These photomicrographs indicate that the coated particles contacting the thin powder-metallurgy molybdenum surfaces were deformed to produce smooth surfaces. However, little deformation is apparent in the surface particles that contact the thicker arc-cast molybdenum. Thus, we conclude that the type of container material contacting the coated particles is important in establishing the smoothness of cermet surfaces.

The photomicrographs shown in figure 6 indicate that molybdenum produced by powder-metallurgy techniques is more useful as a container material than is arc-cast molybdenum. However, other cermets produced in containers that used powder-metallurgy molybdenum for all components also exhibited irregular surfaces. Thus, the difference in the ability of various molybdenum plates to deform surface fuel particles probably is related to small differences in composition or to different amounts of reduction.

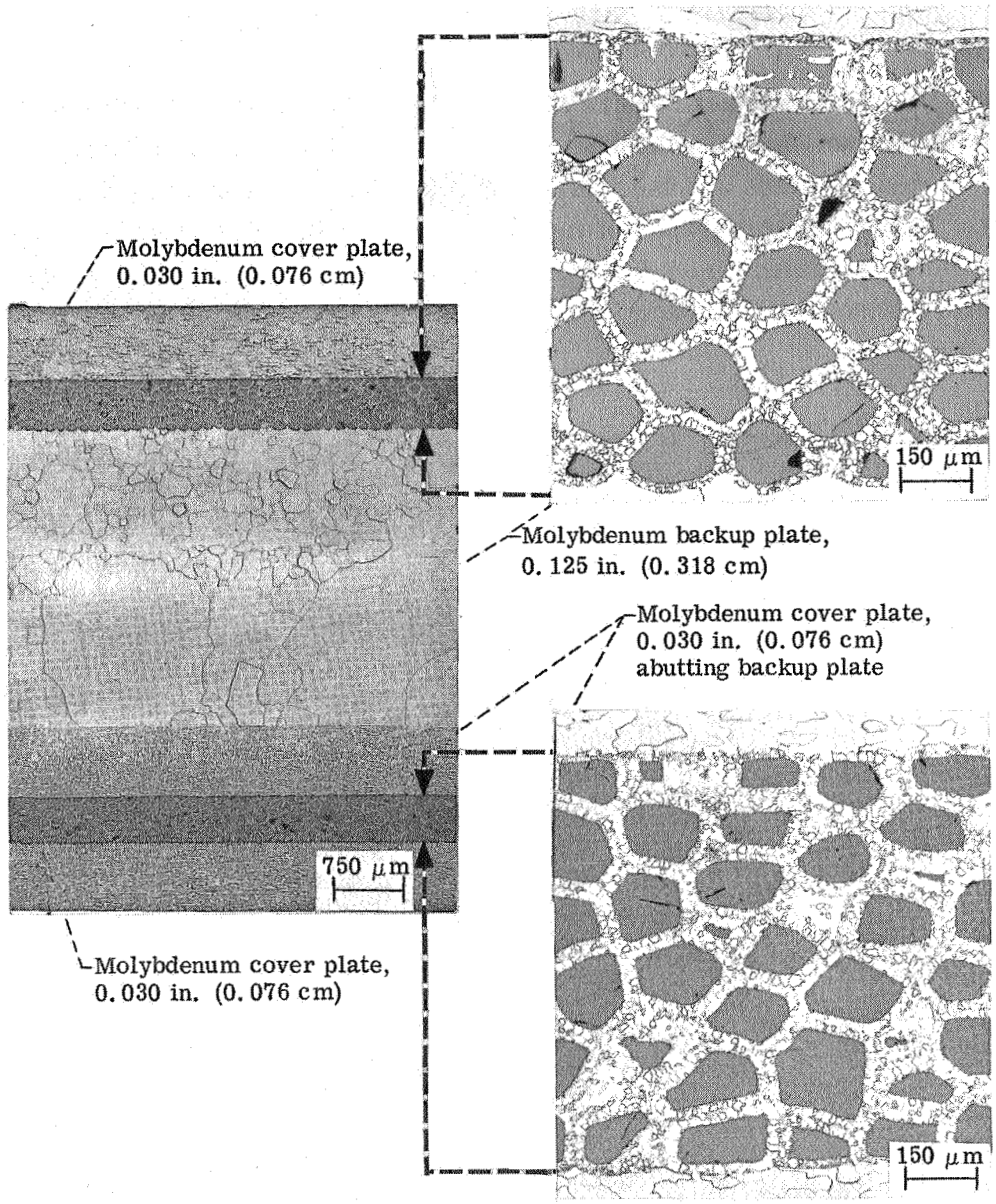
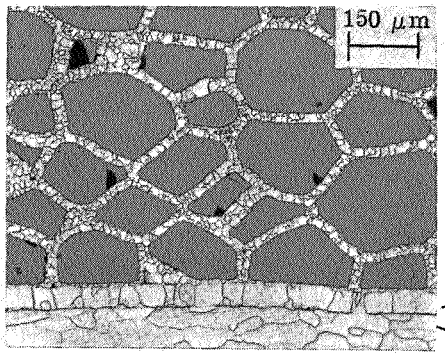


Figure 6. - Effect of container material on uranium dioxide surface-particle deformation of tungsten - 50 volume percent uranium dioxide specimens. Etchant, Murakami's reagent.

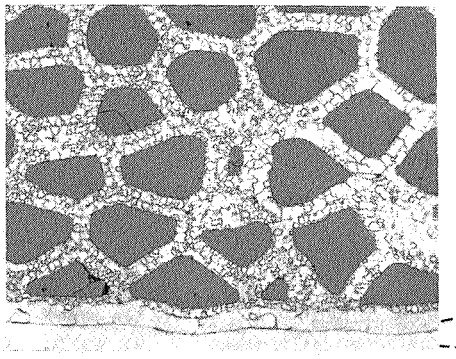
Because of the irregular surfaces that result with the use of unalloyed molybdenum containers, we recommend the use of the TZM alloy for compacting W-UO_2 cermets that require smooth surfaces. The higher elevated temperature strength of this alloy probably accounts for its better resistance to deformation. Thus, other high-strength molybdenum alloys could also probably be used for container materials.

Feasibility of Applying Tungsten Claddings

For W-UO_2 cermets that require an additional thin tungsten cladding on their surfaces, unalloyed molybdenum containers can be used with a tungsten liner interposed between the coated particles and the molybdenum (as shown in container designs of fig. 2(c)). The photomicrographs of figure 7 show that both the tungsten foil and the vapor-deposited-tungsten liners bonded completely to the fuel particle coatings and caused deformation of the surface fuel particles.



(a) Tungsten - 70 volume percent uranium dioxide; vapor-deposited-tungsten liner.



(b) Tungsten - 50 volume percent uranium dioxide; tungsten-foil liner.

Figure 7. - Photomicrographs of integrally clad tungsten - uranium dioxide cermets. Etchant, Murakami's reagent.

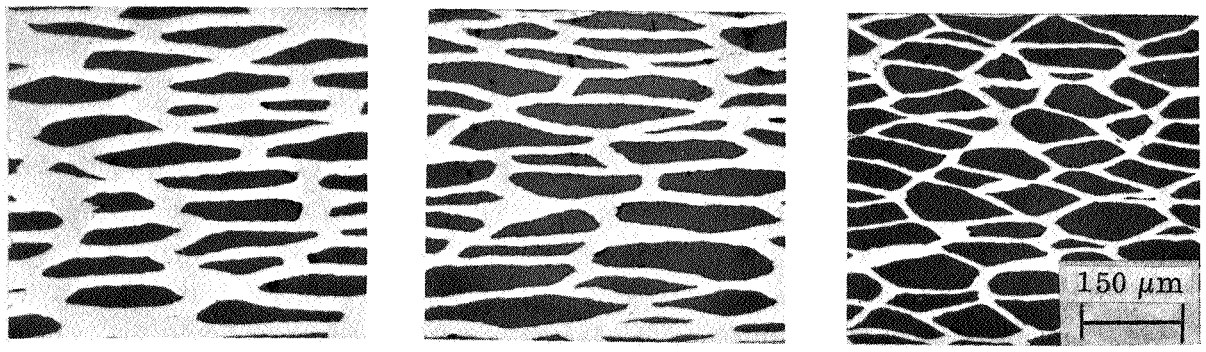
Insufficient work was done to provide a basis for recommending a cladding process. However, the following general observations were made. The vapor-deposited tungsten claddings are more expensive than foil claddings. But the foil claddings are more difficult to handle during assembly operations and may wrinkle. Wrinkled foil will produce a fuel element with a "wavy" surface because, during compaction, the fuel compact will flow to conform to the geometry of the cladding, as shown in figure 7(a).

Although further development of this cladding process is needed, the use of tungsten container liners appears to be an excellent method of applying claddings to the composite as an integral part of the compaction process.

Comparison with Hot-Roll Compaction

To evaluate further the hot isostatically compacted cermets produced in this study, they are compared with similar W-UO_2 cermets fabricated by hot-roll compaction. The latter cermets were produced by Gordon K. Watson of the Lewis Research Center using the method described in reference 2. In the hot-roll compaction method, tungsten-coated UO_2 particles were canned in thin molybdenum containers and hot rolled (at about 3100° F or 1700° C) to compact the particles.

Photomicrographs of cermets produced by each process and containing 30, 50, or 70 volume percent UO_2 are shown in figures 4 and 8. The only significant difference in materials used for these specimens was the original fuel particle size: the hot-roll compacted specimens were produced from 50-micron UO_2 particles (average particle diameter), while the hot isostatically compacted specimens were produced from 110-micron UO_2 particles (average particle diameter). The photomicrographs shown in figures 4 and 8 indicate that both compaction processes produced fully dense cermets with uniform fuel distribution and complete separation of the fuel particles. However, the hot-isostatic-compaction process caused less deformation of the fuel than did the hot-roll-compaction process. The more nearly equiaxed fuel particles resulting from hot isostatic compaction should yield more isotropic properties in these cermets, particularly in thermal



(a) Tungsten - 30 volume percent uranium dioxide.

(b) Tungsten - 50 volume percent uranium dioxide.

(c) Tungsten - 70 volume percent uranium dioxide.

Figure 8. - Photomicrographs of fully dense, hot-roll compacted specimens. Unetched.

conductivity and tensile strength. Also, the tungsten matrix separating the fuel particles remained more uniform in the hot isostatically compacted cermets, particularly at the highest fuel loading.

Another advantage of hot isostatic compaction is that it is probably more applicable to a wide variety of configurations. Hot-roll compaction is limited to plates and other simple geometries that can be rolled. However, hot isostatic compaction has been used to produce relatively complex W-UO_2 configurations (ref. 1). Thus, hot isostatic compaction appears to be the more useful process for producing W-UO_2 fuel elements for space power reactors.

CONCLUSIONS

The following conclusions were drawn from an attempt to develop hot isostatic compaction procedures for producing dense tungsten - uranium dioxide cermets from tungsten-coated uranium dioxide particles:

1. Dense tungsten - uranium dioxide (>98 percent of theoretical) composites containing from 30 to 70 volume percent uranium dioxide were successfully fabricated in plate and rod configurations by hot isostatic compaction at 30 000 psi (217 MN/m^2) and 3100° F (1700° C). All these composites exhibited uniform fuel distribution and complete separation of the fuel particles.

2. Variations in uranium dioxide particle size (in the range of 50- to 110-μm diam) had no appreciable effect on cermet density under the compaction conditions used in this investigation. Also, higher compaction pressures (up to 42 000 psi or 290 MN/m^2) at 3100° F (1700° C) had no appreciable effect on the composite density or microstructures.

3. Surface smoothness of the composites was dependent on the container material used. All cermets canned in a molybdenum alloy (TZM) had smooth surfaces, but the use of unalloyed molybdenum containers yielded varying degrees of surface smoothness. Thus, the higher strength molybdenum alloys are recommended for use as container materials.

4. Thin tungsten claddings were successfully applied to tungsten - uranium dioxide plates and rods as an integral part of the hot isostatic compaction process.

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